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Mini Review

Overview of Electrochemical Method in the Treatment of Municipal Sewage

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The discharge of municipal sewage has been increasing gradually, causing great harm to the ecological environment. The study of municipal sewage treatment has received increased attention. The recent research progress of municipal sewage detection, treatment and reuse methods, are reviewed. However, each method has its own limitations, such as incomplete treatment, high cost and secondary pollution. This review focus on the electrochemical methods and its applications in the treatment of municipal sewage, including contaminant detection, removal of organic matter, nitrogen, phosphorus, microbial, heavy metal, and degradation of drug residues. The results from the present review demonstrate that the electrochemical technology is an advanced method for municipal sewage treatment due to the advantages of easy-operating, environmental protection and strong universality. Moreover, various new electrode materials, such as graphene, precious metals and polymers are innovated continuously, which lay a good foundation for the application of electrochemical method in municipal sewage treatment. However, some problems still exist in the industrialization of technology, comprehensive treatment of pollutants and degradation capacity of individual pollutants. Herein, some suggestions are proposed for further work of electrochemical treatment in municipal wastewater.

Keywords: electrochemical methods; municipal sewage; pollutants; treatment

1. INTRODUCTION

In recent years, the process of urbanization has been accelerating. At the same time, the population of the city has been increasing rapidly [1, 2]. As a result, the discharge of municipal sewage has been increasing year by year, which has caused serious harm to the ecological environment [3]. The researches of municipal sewage treatment have drawn great attention. The municipal sewage mainly comes from the citizen's domestic water, the commercial water, and the municipal unit water. This sewage finally gathers through various types of urban drains. The municipal wastewater must be treated properly, otherwise, it will pose severe risks to the natural environment [4]. Therefore, the effective treatment of municipal sewage has always been the focus of researchers [5]. The number of research papers has reached 6205 in the recent 10 years (2012–2021) from Web of Science using the terms "Municipal sewage," and an increasing trend is shown year by year (Figure 1).

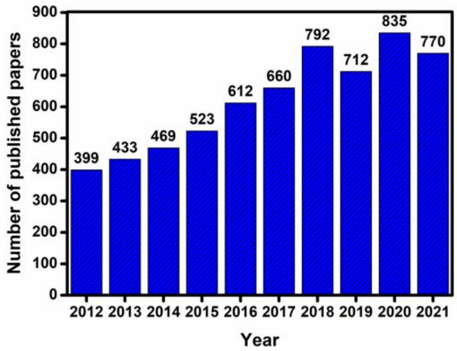


Figure 1. Annual number of published research papers related to Municipal sewage published from 2012 to 2021

The composition of municipal sewage is complex because of its complicated sources [6], including organic pollutants [7], particulate matter, nitrogen, phosphorus, medical residues [8, 9], microorganisms and bacteria [10, 11], etc. However, the composition of municipal sewage is relatively stable compared with the industrial effluents, which brings great convenience to the effective treatment of municipal sewage.

To gain insight into the electrochemical treatment, recent research progress of municipal sewage treatment is reviewed. The focus is placed on the principle, application and progress of electrochemical method. The merits and demerits of electrochemical wastewater treatment are also summarized in this review. Considering the development potential of electrochemistry and the demand of the public, some perspectives are put forward for further work on the electrochemical treatment of municipal wastewater.

2. RESEARCH PROGRESS AND EXISTING PROBLEMS OF MUNICIPAL SEWAGE TREATMENT METHODS

The technology of municipal sewage treatment has been improved constantly in recent years, and many types methods of municipal sewage detection, treatment and reuse has been developed. We have developed considerable detection methods for various pollutants, such as viruses, bacteria, antibiotics, metal particles and other contaminants. For example, Aiqiang Xu used next-generation sequencing to detect enteroviruses in municipal sewage and studied its application in environmental monitoring [11]. Siva Prasad Bitragunta et al. applied inductively coupled plasma mass spectrometry (ICP-MS) technology to quantify titanium and characterize titanium dioxide particles (TNP) [12]. Haiwei Huang et al. identified the biotic and antibiotic resistance genes in municipal wastewater and analyzed their abundance and diversity [8].

In the field of pollutant treatment, many scholars have studied the chemical oxidation method, physical adsorption method, activated sludge method, biological method and constructed wetland method. For instance, in chemical oxidation, Yuan Wei developed one-stage anaerobic ammonium oxidation (Anammox) for nitrogen removal [13]. Xiong Hong-bin et al. used graphene photocatalytic oxidation technology to improve water quality significantly [14]. In physical adsorption, F. C. Kramer et al. applied ceramic nanofiltration membrane to purify municipal wastewater [15]. Z. Sapronova et al. provided an effective method for adsorbing pollutants in municipal wastewater with modified chestnut tree waste [16]. Qian Wu et al. reported the removal of estrogen with activated sludge [17]. In biological method, Jie Xu et al. demonstrated the excellent performance of microalgae technology to remove pollutants in domestic sewage [18]. Thomas et al. proposed to treat municipal wastewater with the algal floway method and evaluated its biomass yield, lipid content and biofuels production [19]. Xia Huang et al. adopted anaerobic forward osmosis membrane bioreactor anomer to treat municipal domestic sewage [20]. A submerged anaerobic membrane bioreactor was adopted to purify low concentration domestic sewage by Jianwei Liu et al.. [21]. Agostina Chiavola and Mei Chen et al. reported the effect of biofilm oxidation reactor on drug degradation [22, 23]. Bin Ji et al. used sequencing batch biofilter composed of fibrous packing and ceramists filter to treat domestic sewage [24]. Biological phosphorus removal from aerobic wastewater was studied by Hongbo Chen et al.. [25] In the field of constructed wetlands, Zhiwei Zhou et al. developed the double-layer substrate formed by covering shale ceramsite on activated alumina in tidal flow constructed wetland for enhancing nitrogen removal from decentralized municipal sewage [26]. Shibao Lu et al. developed anaerobic flow constructed wetlands to remove nitrogen [27]. Tatiane Benvenuti et al. used aquatic plant spathe Typha orientalise to construct floating wetlands to treat municipal sewage [28]. M. Anjos et al. studied the removal of Paraben by duckweed [29].

In addition, some studies focus on the reuse of municipal sewage. For example, Noriatsu Ozaki et al. reported the composting of municipal sewage sludge [30]. Beibei Yan et al. adopted the combination of anaerobic digestion and pyrolysis to treat municipal sewage sludge for recycling organic matter [31].

These methods played an important role in the treatment of municipal sewage at different stages, but each method has its own limitations, such as incomplete treatment, high cost and secondary pollution. The advantages and disadvantages of traditional municipal sewage treatment methods are shown in Figure 2.

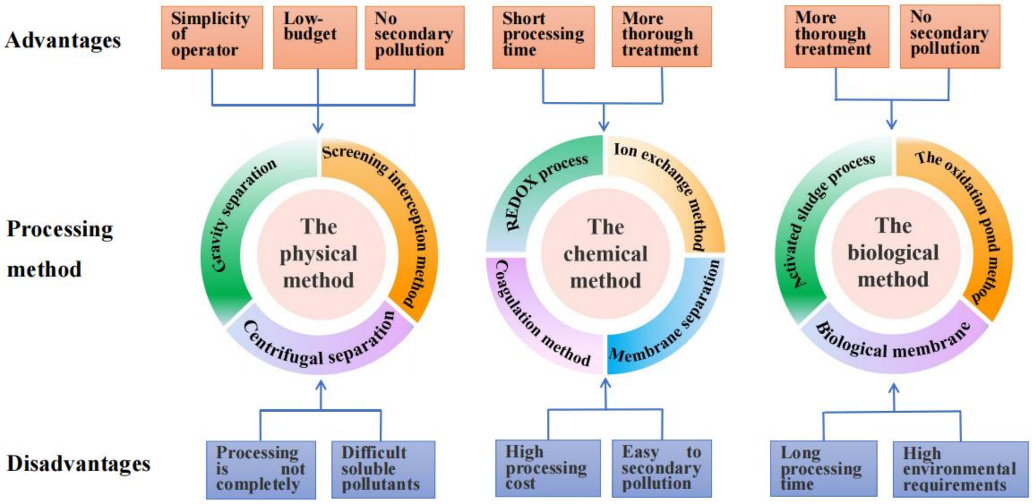


Figure 2. Comparison of advantages and disadvantages of traditional sewage treatment methods

This study introduces a more suitable method for the treatment of municipal sewage, that is, electrochemical method.

3. RESEARCH PROGRESS OF ELECTROCHEMICAL METHODS IN THE TREATMENT OF MUNICIPAL SEWAGE

3.1 Application of electrochemical method in contaminant detection

Electrochemical methods have been used in the detection of municipal wastewater for a long time. Many scholars have conducted extensive studies on various aspects. For the application of electrochemical methods to detect different pollutants, Chantemesse Benoit et al. developed β-lactam electrochemical method based on nitro-cephalosporin to detect antimicrobial resistant escherichia coli, which had advantages in quick, convenient, low cost and convenient carrying [32]. Dingnan Lu et al. detected SARS-CoV-2 in wastewater using electrochemical immunosensors, and the result was favourable [33]. Zhilei Li et al. studied the detection of abiotic stress biomarkers, including various plant hormones, by electrochemical methods [34]. Hassan Karimi-Maleh developed a novel molecularly imprinted electrochemical sensor for the determination of organophosphorus pesticide residues [35]. Krishnapandi Alagumalai et al. synthesized metal silver incorporated carrom coin structured cobalt oxide (Ag-Co3O4 NPs) by co-precipitation method and achieved the electrochemical detection of Cytotoxic Tinidazole [36]. Oliveira, G. et al. developed a portable electrochemical assay for detecting active ricin based on theelectro-oxidation of adenine released from herring sperm DNA substrate by its catalytic action [37]. Sanaz Pilehvar et al. developed aptamer electrochemical biosensor for the detection of organic matter in sewage, and the detection limit had been reached 1×10^{-9} M [38]. In terms of heavy metal detection, Chailapakul Orawon et al. developed microfluidic chip for detecting lead, cadmium, copper and other pollutants rapidly and simultaneously [39]. These studies accomplished the electrochemical detection of different pollutants.

The detection sensitivity has been rapidly improved in recent years. For example, Wee, Youngho et al. prepared a phenol biosensor with high sensitivity by adding carbon nanotubes to the enzyme electrode. The detection sensitivity of phenol was up to 1170 mu A/mM/cm [40]. Yang Liu et al. developed enzyme biosensor, which was based on biochar nanoparticles for the sensitive detection of bisphenol A (BPA). The linear range was from 0.02 µM to 10 µM, and the lowest detection limit was 3.18 nM [41]. Korkut Seyda et al. developed an amperometric biosensor based on gold nanoparticles embedded polypropylene imidazole for highly sensitive detection of urea, and the detection limit reached 36 µM, a relative standard deviation of 2.43% (n = 18) [42]. Tsai Wan-Ting et al. developed an ultrasensitive photonic refractive index sensor based on nano porous silicon to detect heavy metal ions in water. The detection sensitivity of cadmium ion reached 342 nM/RIU and 0.152 ppb [43].

Kaur Ramandeep et al. developed fabric phase adsorption extraction technology to detect trace organic compounds, and the detection limit reached 0.009–0.021 ng/ml [44]. J. Gallardo–Gonzalez et al. developed a real-time potential measurement chip for the realtime detection of pollutants in flowing water by electrochemical method [45]. Makombe Martin et al. developed adsorptive stripping voltammetry for the determination of rare earth elements [46]. Yang Si et al. developed TiO_2 photo assisted electrochemical sensor for detecting Bisphenol A (BPA) [47], providing a powerful sensor for the detection of pollutants in water. The development of these different detection methods provides more approaches to detect pollutants in municipal sewage and improves the ability of detection.

The types of pollutants are more abundant. The detection sensitivity is higher, and the detection forms are diversified.

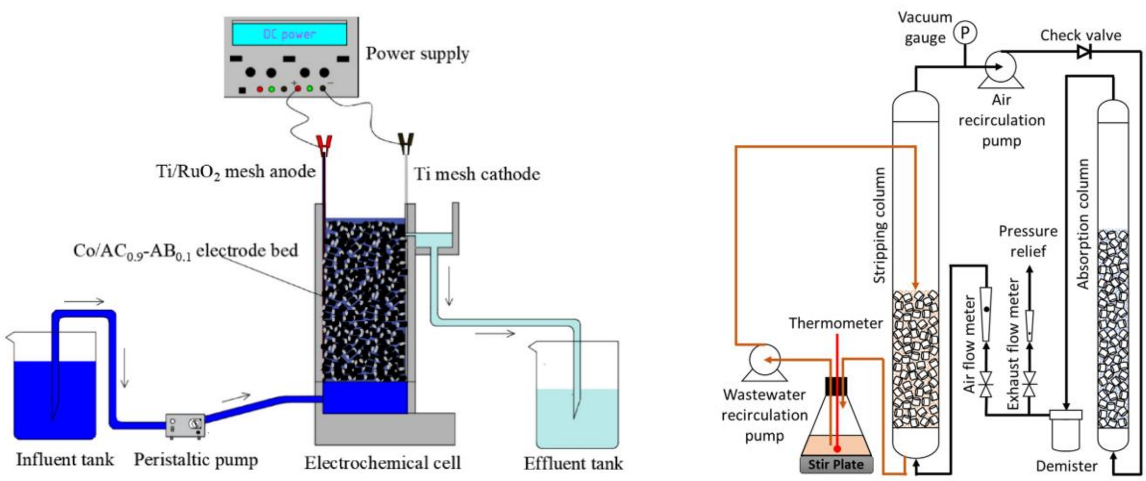
3.2 Research progress of electrochemical methods in organic matter removal

The electrochemical catalytic degradation of organic compounds is the direct oxidation of organic compounds on the anode or generation of hydroxyl radical, ozone and other oxidants on the anode. For example, Rajamohan Natarajan et al. synthesized a new type of natural coagulant with pea as raw material. The chemical oxygen demand (COD) in municipal sewage was effectively removed in the reactor by using aluminum as the electrode, thereby confirming the feasibility of parallel plate electroflocculation reactor with natural coagulant for municipal sewage treatment [48]. Zheng Wentian et al. established an electrochemical filtration system using carbon nanotubes (CNT) and nano zero valent copper (NZVC) composite materials as high-performance catalysts, electrodes, and filter media. This system achieved continuous degradation of organic pollutants in a wide range of pH values, which provided an effective method for the degradation of complex organic compounds in complex water environments [49]. Based on the combination of nuclear magnetic resonance (NMR) and electrochemistry, Xiong Liangliang et al. developed a method for the adsorption and radical oxidation of bisphenol by graphite electrode in homogeneous Fe (III) / persulfate system, which realized the efficient degradation of organic compounds [50]. The electrochemical flow cell direct coupled with mass spectrometry technology was used to catalyse the oxidation of carbendazim (CBZ) fungicide by

Temgoua Ranil C. T. et al., which proved that the electrochemical method combined with other methods has a perfect application prospect [51]. For the research of the mechanism, Yang Deng et al. clarified the changes in the properties (complexing ability, aromaticity and mobility) of humic substances in the process of electrochemical oxidation, and explained the mechanism of the changes, which provided the basis for further understanding and evaluating the electrochemical degradation process of humus [52].

3.3 Research progress of electrochemical methods in nitrogen and phosphorus removal

In recent years, the removal of nitrogen and phosphorus from wastewater by electrochemical method is also a research hotspot. The removal rate of nitrogen and phosphorus has increased, and some new electrochemical methods have emerged. For instance, Yang Lei et al. developed an electrochemical method for phosphorus removal and recovery by locally increasing the pH near the cathode [53]. The addition of chemicals to increase the pH of the bulk solution and the further separation step during traditional chemical precipitation process was avoided by Yang Lei et al [53]. Wenjian Ye et al. used cobalt as the catalyst, active carbon (AC) as the carrier, and acetylene black (AB) as the conductor to establish a composite particle electrode to remove TN, and the removal rate reached 95% [54] (Figure 3). Mahamalage Kusumitha Perera et al. developed a continuous-flow reactor recovery of 89% and 97% of average total N & P from municipal sewage by precipitation of $Ca_3(PO_4)_2$ and stripping of $NH_3(g)$, following electrochemical pH shifting, termed electrolyte modulation [55] (Figure 3). The pollution is removed, and the resources are recycled. Qiao Sen et al. developed an electrochemical method to remove nitrate by coupling anammox with autotrophic denitrification. The removal rate is as high as 99.1%, which is the highest in the literature [56]. Tejedor–Sanz Sara et al. developed a new electrochemical nitrogen removal method and constructed microbial electrochemical fluidized bed reactors (ME-FBR) to promote bacteria electrode interaction. This study was the first to propose that fluidized bed could remove 98% of ammonia nitrogen as cathode reaction [57].



(a) (b)

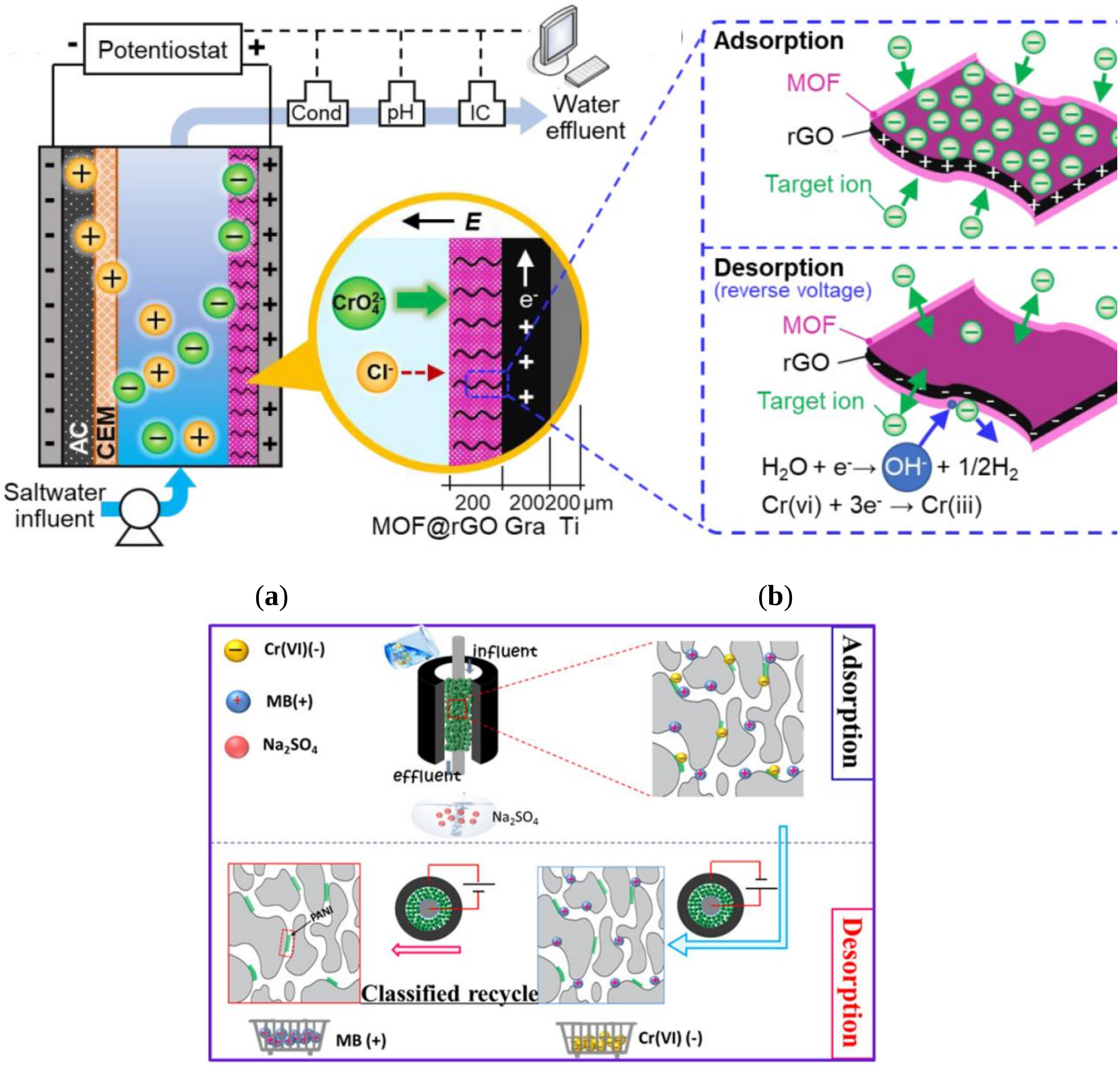
Figure 3. (a) Schematic view of the continuous 3D electrochemical reactor [54] Copyright © 2020, Elsevier; (b) Continuous-flow nutrient recovery apparatus [55] Copyright © 2020, Elsevier.

3.4 Research progress of electrochemical methods in removal of microbial indicators

Electrochemical method has few reports on microbial removal, but some scholars have conducted indepth research. For example, in the early days, Voccia, S. et al. electrodeposited metal silver / polymer composite layers on conductive substrates, and the silver coating could effectively inhibit the growth of gram-negative Escherichia coli [58]. Subsequently, Javier et al. applied conductive diamond ultrasonic electrochemistry method to disinfect municipal sewage and effectively inhibited E. coli [59]. Qian Lei et al. established an electrochemical dynamic membrane filtration (EDMF) system to simultaneously realize solid–liquid separation and pollution degradation. The degradation of bacteria achieved 100% under the condition of 2.5 V low voltage [60], providing an effective method for microbial degradation in wastewater by electrochemical method. Ma Qiang et al. used hemin / graphite felt (GF) composite electrode to kill E. coli. The killing rate reached 99%, and the disinfection of drinking water was realized at low potential (0.6 V) without chlorine [61]. Thus far, electrochemical methods have shown great advantages in sterilization and anti-virus.

3.5 Research progress of electrochemical methods in heavy metal removal

In recent years, studies on the removal of heavy metals by electrochemical methods. For example, Kuichang Zuo et al. developed a new electrochemical method based on hybrid MOF@rGO for high selective degradation of Cr (VI) with high affinity adsorbent electrode [62](Figure 4). Jiancheng Shu et al. developed a new electrochemical method for efficient removal of Mn^{2+} and NH^{4+}-N simultaneously with Cu plate as cathode and the removal efficiencies were 99.1% and 92.9% respectively [63]. Jie Ding et al. developed an electrochemical assisted desorption method to enhance polyaniline modified spherical resin (PANI@PS). The maximum recoveries of Cr (VI) and dyes were 90.3% and 91.9% [64](Figure 4), respectively. This study is greatly important for the introduction of new adsorption composite materials for simultaneous removal and fractionation of specific coexisting pollutants. Xu Jinwei et al. developed an asymmetric AC electrochemical method to remove different heavy metal (copper, lead, and cadmium) at different initial concentrations (100–10000 ppm) to reach the relevant regulatory level [65], which has strong practicability.



(**a**) (**b**)

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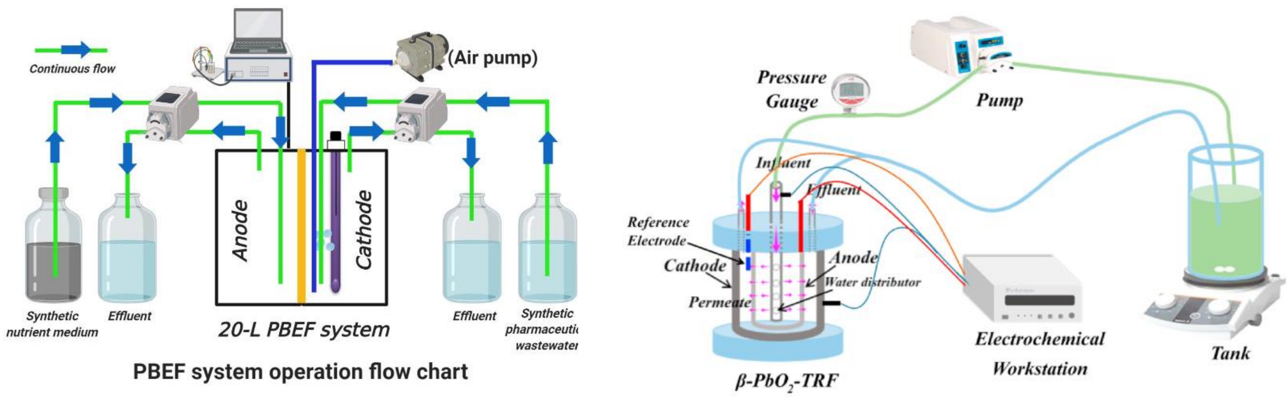
Figure 4. Schematic showing the use of MOF@rGO in an asymmetric electrosorption system for selective removal of Cr(VI) [62] Copyright © 2020, American Chemical Society and Cr(VI)(−) and MB(+) desorption [64] Copyright © 2019, Elsevier. (a) Schematic of MOF@rGO as a selective electrode in the asymmetric electro sorption system. Gra: graphite sheet. Ti: titanium sheet [62] Copyright © 2020, American Chemical Society; (b) Enhanced adsorption and desorption/reduction of Cr(VI) in a charge/reversed voltage cycle [62] Copyright © 2020, American Chemical Society; (c) Schematic of Cr (VI)(−) and MB (+) desorption [64] Copyright © 2019, Elsevier.

3.6 Research progress of electrochemical methods in degradation of drug residues

Drug residues have the characteristics of high toxicity and difficult degradation, which has always been the difficulty of wastewater treatment. However, the electrochemical method has strong preponderance in the degradation of drug residues [66]. Many scholars have conducted considerable in-depth research and have achieved good results. For example, Rusen Zou et al. developed a microbial electrochemical ultraviolet photolysis cell that could effectively eliminate carbamazepine and provided a new method for the treatment of medical residual wastewater [67] (Figure 5). Kui Yang et al. developed an ultrahigh throughput tubular reactive filter with β-PbO_2 electrocatalytic layer that could remove norfloxacin and sulfamethoxazole at trace level concentration in wastewater, requiring only 2.0–5.4 s [68] (Figure 5). Chong Wang et al. studied the Ti/SnO_2-Sb/La-PbO_2 anode electrochemical degrade antibiotic enrofloxacin, and their kinetics, mechanism, and toxicity evaluation [69]. Wei Zhang et al. used graphite felt electrode to degrade a typical ionic compound pantothenic acid and the degradation rate was close to 100% in two hours [70]. Yassine Ouarda et al. used electrochemical method by boron-doped diamond (NB/BDD) anode and titanium (Ti) cathode to remove 12 mixed drug residues (caffeine, cihydrocarbamazepine and desvenlafaxine). After 120 minutes, the removal rate reached 100% [71].



(**a**) (**b**)

Figure 5. This is (a) Flowchart of 20 L MEUC system [67] Copyright © 2020, Elsevier; (b) Schematic of the tubular reactive filter system consisting of a perforated stainless-steel pipe as the inner cathode and the water distributor, a stainless-steel pipe as the outer cathode, a pump, an electrochemical workstation, and a solution tank [68] Copyright © 2020, Elsevier.

4. CONCLUSIONS

Some methods of municipal sewage treatment are systematically studied in recent years. Researches focus on the electrochemical method and its application in municipal sewage treatment. Electrochemical method is widely used in the detection of various pollutants, degradation of organic matter, removal of nitrogen, phosphorus and heavy metals, microbial killing and degradation of residual drugs in wastewater treatment due to its advantages of simple operation, green environmental protection and strong universality. The removal rate of ammonia nitrogen, some heavy metals, bacteria and drug residues can reach 98%–100%. Great progress has been made in the selection of electrochemical methods, such as molecularly imprinted method, asymmetric alternating current electrochemical method, adsorptive stripping voltammetry, microbial electrochemical fluidized bed method and microbial electrochemical ultraviolet photosynthesis cell. The continuous innovation of polymer and other electrode materials or composite electrode materials has laid a good foundation for the application of electrochemical methods in municipal sewage treatment.

However, many problems remain unresolved. First, the actual application of technology is still poor. Transforming “advanced technology” into “application technology” is still a serious challenge. Second, degrading multiple pollutants simultaneously is still difficult by the same electrochemical method, and the simultaneous degradation of complex polluted water is still an urgent problem to solve. Third, although electrochemistry has a favorable effect on the removal of heavy metals, drug residues, nitrogen and phosphorus, the complete degradation of microorganisms and organic matter is not ideal, and further improvement is still desired. Therefore, in order to give full play to the advantages of electrochemical method and better apply it to municipal sewage treatment, further work should be carried out in the following aspects:

To further increase the pilot scale and pilot scale tests to lay the foundation for further industrialization based on the good effect of electrochemical method laboratory in treating municipal sewage.

To further develop electrochemical methods for simultaneous degradation of multiple pollutants in complex water and explore their coupling mechanism.

To further improve electrode materials or develop new electrochemical methods to explore the mechanism of organic matter and microbial degradation to thoroughly degrade organic matter and microorganisms.

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